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Paul D. Ronney
*Lewis Research Center
Cleveland, Ohio*

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EFFECT OF GRAVITY ON HALOGENATED HYDROCARBON FLAME RETARDANT EFFECTIVENESS

Paul D. Ronney*
National Aeronautics and Space Administration
Lewis Research Center
Cleveland, Ohio 44135 U.S.A.

Abstract

E-2251
Flammability limits, burning velocities, and minimum ignition energies under initially quiescent conditions were measured for stoichiometric and fuel-lean methane-, ethane-, and propane-air mixtures containing varying concentrations of Halon 1301. The characteristics of near-limit flames were strongly affected by fuel type but not Halon concentration. The conclusions were that the mechanism of the flammability limits was affected by fuel type but not Halon concentration, that the zero-g flammability limit is probably related to a stability criterion which is mostly dependent on the molecular diffusion characteristics of the reactant gases and is mostly independent of chemical kinetics, and that the one-g upward flammability and ignition limits provide adequate criteria for safety at one-g and zero-g for both uninhibited and inhibited mixtures.

Background

It is well known that gravity affects the properties of many types of flames.^{1,2} The study of these effects is of value to the spacecraft designer, as it is important to know if potentially flammable materials determined to be fire-safe at normal earth gravity (one-g) are also fire-safe at zero-gravity (zero-g) and if fire extinguishment systems tested at one-g are also effective at zero-g. The U. S. Space Shuttle orbiters use a fire extinguishment system employing Halon 1301 (CF_3Br) as a flame retardant yet no information is available on the effectiveness of this or any other flame retardant at zero-g. Flame retardants are commonly tested by measuring the flammability limits, burning velocities, and minimum ignition energies of premixed gas flames, usually hydrocarbon-air, mixed with varying concentrations of the retardant.³⁻⁵ Thus, such tests performed at zero-g would have practical value.

A thorough discussion of the mechanism of flame inhibition is beyond the scope of the current work. Many reviews appear in the literature; a listing of these is given in Ref. 6.

Recent studies⁷⁻⁹ have shown that there are considerable differences between premixed gas flame propagation at one-g and zero-g near the flammability limits. In particular, it has been found that at zero-g, for mixtures near the zero-g flammability limit, an unstable mode of flame propagation (known as Self-Extinguishing Flames, or SEFs) occurs which is not observed at one-g. Factors thought to cause flammability and/or ignition limits at one-g, such as heat losses, "flame stretch",¹⁰ pressure rise in closed-vessel flames, and insufficient spark energy, do not seem to

account for the phenomena observed at zero-g. The cause of SEFs has not been determined but it appears to be related to aspects of chemical kinetics or transport properties which are masked at one-g by natural convection. Small quantities of flame inhibitors can alter the flame kinetics without significantly altering the transport properties.^{11,12} Hence, a study of the effect of flame inhibitors at zero-g may provide new insight into the causes of zero-g flammability limits and SEFs.

Thus, a study of the effect of flame inhibitors on premixed gas flames at one-g and zero-g would have both practical and scientific value. In the current study, Halon 1301 is employed because of its widespread use in practical systems. A large closed vessel is used to permit testing over a range of pressures without significant heat loss to the apparatus and to allow comparison with previous zero-g experiments.^{7-9,13}

Experimental Apparatus and Procedures

The experimental apparatus (Fig. 1) has been described previously.⁷ It consisted of a combustion vessel, motion picture camera, spark generator, spark energy computer, event timer and sequencer, and batteries, all mounted in an aluminum framework. The combustion vessel was an aluminum cylinder, 25 cm in diameter and 25 cm long with Plexiglas windows at both ends, fitted with radially opposed spark electrodes having 0.35 mm diameter tips. Standard optical photography was used. The same apparatus was used for one-g and zero-g tests. The zero-g tests were performed during 2.2 sec of free-fall in a drop tower. The methods of determining the gas mixture ratios, flammability limits, burning velocities, and minimum ignition energies are identical to those of previous studies using the same apparatus.⁷⁻⁹

Results

Qualitative

Flames in uninhibited mixtures were pale blue in color and light was visible only from the leading edge of the flame front. The inhibited flames were bright yellow-orange in color, even in mixtures containing only a small amount of Halon, and the entire volume of burned gas continued to glow until the flame consumed all of the available fuel or extinguished. Flames in inhibited propane mixtures had a distinct blue edge, probably due to the greater diffusivity of oxygen molecules relative to fuel molecules in propane flames; this would lead to preferential diffusion of oxygen molecules into the burned gas and thus a richer mixture at the leading edge of the flame front. In uninhibited mixtures, SEF flame fronts propagated outward at a continually decreasing rate and extinguished suddenly before reaching a propagation rate of zero. In inhibited

*National Research Council Resident Research Associate; Member AIAA.

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mixtures, SEF flame fronts propagated outward in a manner similar to that of SEFs in uninhibited mixtures, reached a maximum radius, and retreated inward, becoming dimmer and more diffuse before disappearing. The rate of retreat of the flame front appeared to correspond to that expected from simple thermal conduction in a nonreacting gas. This supports the assertion⁸ that in SEFs, chemical reaction occurs mostly normally up to near the extinction point, at which point the reaction rate decreases sharply and the hot gas kernel decays by thermal conduction.

For mixtures with a burning velocity (S_u) greater than 15 cm/sec, flame fronts were mostly spherical at one-g and zero-g, hence values of S_u could be calculated from either one-g or zero-g data. In these cases there was little difference between the one-g and zero-g values. For slow burning mixtures (i.e., for mixtures with low equivalence ratios) at one-g, natural convection caused severe flame front distortion which led to the formation of mushroom-shaped flames.⁶ For still slower burning mixtures, flames were small "wisps" which rose to the top of the vessel and extinguished. As the equivalence ratio (ϕ) was decreased, the transitions in flame behavior were gradual. For slow burning mixtures at zero-g, flame fronts were spherical except for the appearance of lobes in methane flames, probably due to preferential diffusion of fuel molecules into the burned gas.¹⁰ Hence, for slow burning mixtures, values of S_u could be obtained only from zero-g data. At the zero-g flammability limit, there was an abrupt transition from normal flames to SEFs.

The combustion products of inhibited flames were highly corrosive, probably due to the formation of HF, HBr, and Br_2 from CF_3Br . The products may also have been toxic.

Quantitative

Flammability Limits. Figure 2 shows the effect of Halon concentration on methane-air flammability limits. At high Halon concentrations, the zero-g limit is closer to the one-g upward limit, thus Halon seems to be slightly less effective at zero-g in high concentrations. A Halon concentration of about 6 percent rendered methane-air mixtures nonflammable under all conditions tested. The one-g results are in good agreement with those previously reported.¹⁴

The effect of fuel type, characterized by molecular weight, on flammability limits is shown in Fig. 3. For methane and ethane, the zero-g limit is between the one-g upward and downward limits but for propane the zero-g limit is richer than either of the one-g limits. For rich propane-air mixtures, it has been found¹³ that the zero-g limit is between the two one-g limits, hence lean propane-air mixtures appear to be an unusual case. Figure 3 also shows that the relative ranking of these limits is unaffected by the Halon concentration. It is significant that the equivalence ratio at the zero-g flammability limit (ϕ_{f1}) is the same for methane and propane for both uninhibited ($\phi_{f1} = 0.531$) and inhibited ($\phi_{f1} = 0.759$) mixtures. Ethane has a lower ϕ_{f1} ; and the difference between ϕ_{f1} for ethane and the other fuels is smaller for inhibited mixtures.

Figure 4 shows the effect of initial pressure on methane-air flammability limits. For uninhibited mixtures, the flammable range expands with decreasing pressure. For inhibited mixtures, the flammable range expands with decreasing pressure except in the case of the zero-g and one-g upward limits at high pressures. Thus, Halon loses some of its effectiveness at high pressures. The relative ranking of the one-g upward, one-g downward, and zero-g limits is not affected by pressure. The results for uninhibited flames are in good agreement with those previously reported.¹⁵

Burning velocities. Figure 5 shows the characteristics of flame development in zero-g limit flames. The time scale for flame development is seen to decrease with increasing fuel molecular weight. It is unusual that for ethane and propane, the propagation speed decreases after ignition (similar to methane) but then increases before attaining a steady value. It has been observed¹⁶ that flames in lean propane-air mixtures exhibit an increasing propagation speed after ignition, but an initial decrease followed by an increase does not appear to have been reported previously. This behavior was found only for mixtures very near the zero-g flammability limit; if ϕ was increased by 0.02 or more from the limit mixture this behavior was not observed.

Figures 6 and 7 show the effect of Halon concentration and equivalence ratio on burning velocities. Figure 6 shows that Halon is less effective as a burning velocity reducer in high concentrations if the slope of plots of S_u versus Halon concentration is used as a criterion, however if this slope is normalized by $1/S_u$, which may result in a better criterion, the effectiveness increases at high concentrations. The slopes of the curves in Fig. 6 at zero Halon concentration compare favorably with those previously reported.⁴ Figure 7 shows that the plots of S_u versus ϕ are almost straight lines which are truncated at the zero-g flammability limit except for the case of intermediate Halon concentrations (2 to 3 percent) where there is some curvature in these plots, the slope being greater for lower values of ϕ .

Figures 8 and 9 show the effect of fuel type on burning velocities in uninhibited and inhibited mixtures, respectively. The addition of Halon causes a decrease in S_u but little change in the characteristics of the curves. Values of S_u are similar for ethane and propane but lower for methane. The plots are practically straight lines except for inhibited methane mixtures. The values of S_u shown in Fig. 8 are in fair agreement with those reported previously.¹⁷

Figures 10 and 11 show the effect of initial pressure on burning velocities in uninhibited and inhibited mixtures, respectively. For the inhibited flames there is less spread among values of S_u at different pressures. The curve corresponding to 1500 torr is somewhat unusual in that it crosses the 760 torr curve, although this might have been expected considering the effect of pressure on flammability limits in inhibited mixtures (cf. Fig. 4).

Figures 7 to 11 show that, except for ethane, the zero-g limit burning velocities ($S_{u,lim}$) are practically unaffected by the Halon concentration.

The data are summarized in Tables 1 to 3. The increase in $S_{u,lim}$ for inhibited ethane-air mixtures might have been expected since ϕ_{fl} for ethane is lower than that of methane or propane and the difference is smaller for the inhibited mixtures (cf. Fig. 3).

Minimum Ignition Energies and SEFs.

Figure 12 shows the minimum ignition energies of normal flames at one-g and zero-g and the characteristics of SEFs for inhibited methane-air mixtures. Corresponding figures for ethane and propane are not shown because these fuels did not exhibit significant SEF propagation. The upper solid curve of Fig. 12 is the minimum ignition energy for flame propagation throughout the vessel at zero-g. The lower solid curve is the minimum ignition energy at one-g. The dashed curves represent contours of constant extinguishment radius (the maximum radius reached by the flame front as measured from the film records) for SEFs. These dashed curves were obtained by mapping extinguishment radii for varying combinations of ϕ and spark energy and drawing approximate best-fit curves through the data. The numbers in parenthesis on the dashed curves are estimates of the chemical energy released by flame kernels of the indicated size.

Figure 12 shows that the minimum ignition energies of methane-air mixtures are the same at one-g and zero-g except for mixtures near the zero-g flammability limit or leaner, where the energy is greater at zero-g. The same effect was found with ethane- and propane-air mixtures.

Figure 12 also shows that the estimated chemical energy release in SEFs can be orders of magnitude higher than the initial spark energy. The highest ratio found was about 20 000. It is surprising that a developing flame kernel can "remember" after releasing 20 000 times its initial spark energy whether it is an SEF, in which case it extinguishes, or a normal flame, in which case it propagates to the limits of the vessel. By comparison, for ethane and propane the maximum ratio of chemical energy release to the initial spark energy was about 15. It has been shown^{7,9} that these low ratios probably correspond to common non-igniting flame kernels,¹⁰ hence for these fuels it appears that SEFs do not occur and common non-ignition behavior is observed instead. A more thorough discussion of SEFs appears elsewhere.^{7,9} Figure 12 is very similar to the corresponding figure for uninhibited methane-air flames,⁹ hence it appears that the characteristics of ignition and SEF propagation are mostly independent of the Halon concentration. It was found, however, that the characteristic time scale of SEFs was about 25 percent faster in inhibited flames. In both cases the flame front radius was found to increase in proportion to the square root of the time lapse from ignition.

Flames at zero-g which were not spherically symmetric were found for mixtures very near the zero-g flammability limit. Such flames have been observed before,^{7,9} but not to the extent found in this work in inhibited flames. Many flame shapes were found; some flames broke up and extinguished within the volume of the combustion vessel and others reignited into flames which consumed

all of the fuel available. An example of the latter case is shown in Fig. 13. A plot of flame radius versus time for this flame until near the time of breakup (not shown) is indistinguishable from the same plots for SEFs under similar conditions. At the time of breakup, the flame extinguished except at the "top" and "bottom" (such distinctions are meaningless at zero-g), reignited, and the two flame kernels assumed a steady propagation rate about equal to that which would be expected of a normal flame in this mixture. The most likely cause of these nonsymmetric flames would appear to be random disturbances caused by the spark discharge aerodynamics; Fig. 12 shows that disturbances are able to propagate furthest from the ignition source for combinations of ϕ and spark energy very near the zero-g minimum ignition energy curve, which is where the nonsymmetric flames were found.

Discussion

It was found in this work that the characteristics of zero-g limit flames and SEFs are strongly affected by fuel type but not Halon concentration. In order to interpret this behavior, let us consider which of the major processes which occur in premixed gas flames, namely chemical kinetics (i.e., reaction rates and mechanism), molecular diffusion, and thermal conduction, might have been affected by a change in fuel type and/or Halon concentration. Chemical kinetics are in general affected by fuel type but the fuels used in this investigation are chemically similar, hence the difference in kinetics between these fuels is small, particularly in lean mixtures.¹⁰ In contrast, chemical kinetics are strongly affected by the addition of Halon.^{11,12} Diffusion of fuel molecules into the burned gas is affected by fuel type because of differences in molecular weight but diffusion in the opposite direction is probably not affected because the combustion products of the fuels tested are similar. Halon and the products of Halon-inhibited combustion except HF have molecular weights which are probably too high to exhibit significant diffusion compared to other species present. Thermal conduction was probably not affected by fuel type or Halon concentration because all gas mixtures tested were at least 86 percent air.

Thus, in this work it appears that a change in fuel type affected mainly the diffusion characteristics of the unburned gas mixture and a change in Halon concentration affected mainly the chemical reaction rates and mechanism. Only fuel type was found to have a significant effect on the characteristics of limit flames and SEFs, hence it is likely that the mechanisms of these phenomena are related mostly to the diffusion characteristics of the unburned gases. If chemical kinetics were an important factor, the Halon concentration probably would have had more effect on the characteristics of limit flames and SEFs. Thus, the flame front instability found at zero-g which is believed⁷⁻⁹ to be responsible for these phenomena appears to be related mostly to diffusion processes. Preferential diffusion of fuel or oxygen into the burned gases, which are thought^{10,19} to cause cellular flames such as those observed in

this work in methane-air mixtures, might be a diffusion process capable of causing zero-g flammability limits and SEFs, although cellular structure did not appear to cause breakup of near-limit normal flames nor did cells appear in SEFs or flames in ethane-and propane-air mixtures.

It has been shown^{7,9} that radiant heat loss does not seem able to account for zero-g flammability limits and SEFs; further evidence can be seen in this work. Theories which relate flammability limits to heat loss^{18,19} predict that an increase in the volumetric heat loss rate leads to an increase in limit burning velocity and that a plot of S_u versus ϕ should approach an infinite slope at a limit caused by heat loss. These predictions have been verified experimentally.^{20,21} In this investigation, the addition of Halon greatly increased the flame luminosity and thus the volumetric radiant heat loss rate, yet Tables 1 to 3 show that $S_{u,lim}$ was unaffected by Halon concentration (except for ethane) and Figs. 7 to 11 show that $ds_u/d\phi$ is finite at the limit.

It is interesting that while SEFs seem to occur only in methane (and perhaps other fuels with a molecular weight less than oxygen), the other characteristics of the zero-g flammability limits are similar for all fuels tested. This indicates that the phenomena responsible for the zero-g flammability limits may be different from those responsible for SEFs.

Figures 2 to 4 and 12 show that flame propagation occurs over a wider range of conditions at one-g (for upward propagation) than at zero-g. Thus, the one-g flammability and ignition limits provide a conservative indicator of fire safety at zero-g for the conditions tested. Since spacecraft generally must also be fire-safe at one-g, the one-g upward limits are proper criteria for safety considerations.

In order to obtain a better understanding of zero-g flammability limits and SEFs, further experiments using other fuel/oxidizer combinations is recommended. Tests with hydrogen would determine if SEFs can occur in other fuels lighter than oxygen. If flammability limits and SEFs are dependent on relative diffusion of fuel and oxidant and not on chemical kinetics, then results of tests using N_2O (molecular weight 44) as an oxidizer and ethane, propane and butane fuels (molecular weights of 30, 44, 58) would be similar to the results of this investigation, even though the chemical kinetics are different, because of the similarities in relative diffusion. Additional flame temperature profile measurements at zero-g, particularly in these new fuel/oxidizer combinations, are advisable to allow better understanding of the structure of near-limit flames and SEFs.

Acknowledgments

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TABLE I. - EFFECT OF
HALON CONCENTRATION
ON ZERO-G LIMIT
BURNING VELOCITIES
IN METHANE-AIR
MIXTURES AT 760 torr
INITIAL PRESSURE

| Halon concentration, percent | $S_{u,lim}$, cm/sec |
|------------------------------------|-------------------------|
| 0.00 | 1.47 |
| 1.00 | 1.76 |
| 2.00 | 1.43 |
| 3.00 | 1.22 |
| 4.00 | 1.38 |
| 5.00 | 1.54 |
| 5.92 | 1.53 |

TABLE II. - EFFECT OF FUEL TYPE ON
ZERO-G LIMIT BURNING VELOCITIES IN
UNINHIBITED AND INHIBITED MIXTURES
AT 760 torr INITIAL PRESSURE

| Fuel | $S_{u,lim}$, cm/sec | |
|---------|----------------------|-----------------|
| | 0 percent Halon | 2 percent Halon |
| Methane | 1.47 | 1.43 |
| Ethane | 2.58 | 3.58 |
| Propane | 5.30 | 5.47 |

TABLE III. - EFFECT OF INITIAL PRESSURE
ON ZERO-G LIMIT BURNING VELOCITIES
IN UNINHIBITED AND INHIBITED
METHANE-AIR MIXTURES

| Pressure, torr | $S_{u,lim}$, cm/sec | |
|-------------------|----------------------|-----------------|
| | 0 percent Halon | 2 percent Halon |
| 1500 | 1.05 | 1.02 |
| 760 | 1.47 | 1.43 |
| 250 | 2.02 | 1.97 |
| 100 | 2.80 | 2.65 |
| 50 | 3.67 | 4.08 |

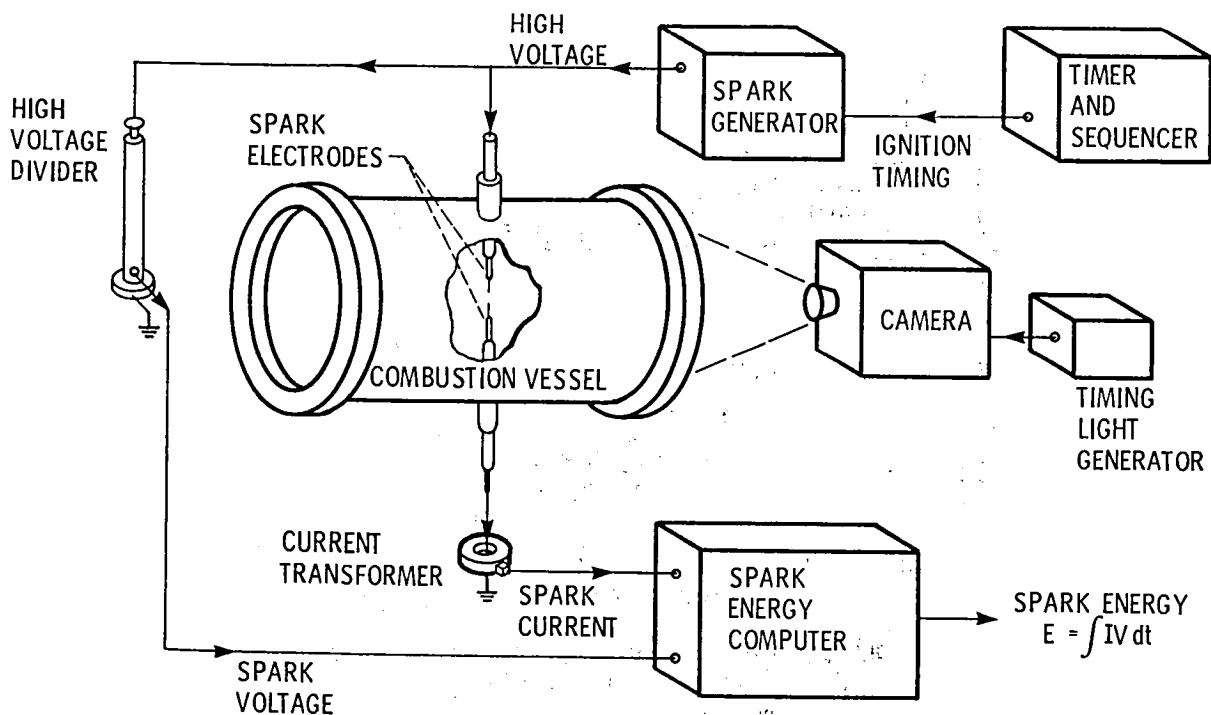


Figure 1. - Experimental apparatus block diagram.

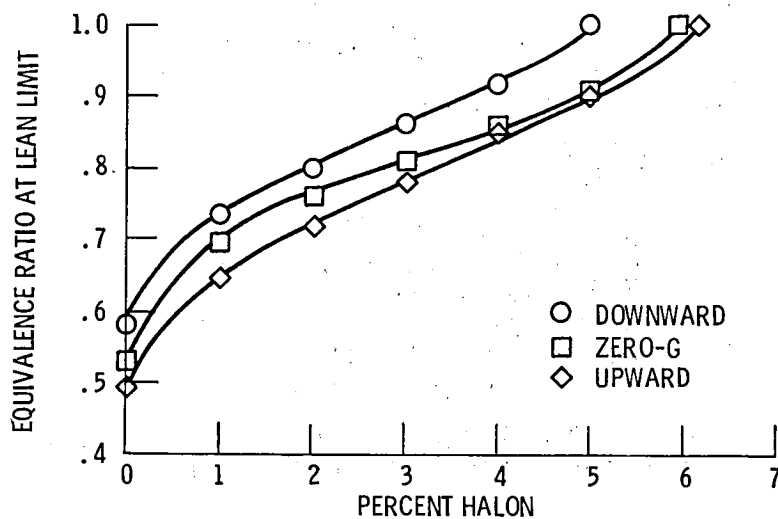


Figure 2. - Effect of Halon concentration on methane-air flammability limits at 760 torr initial pressure.

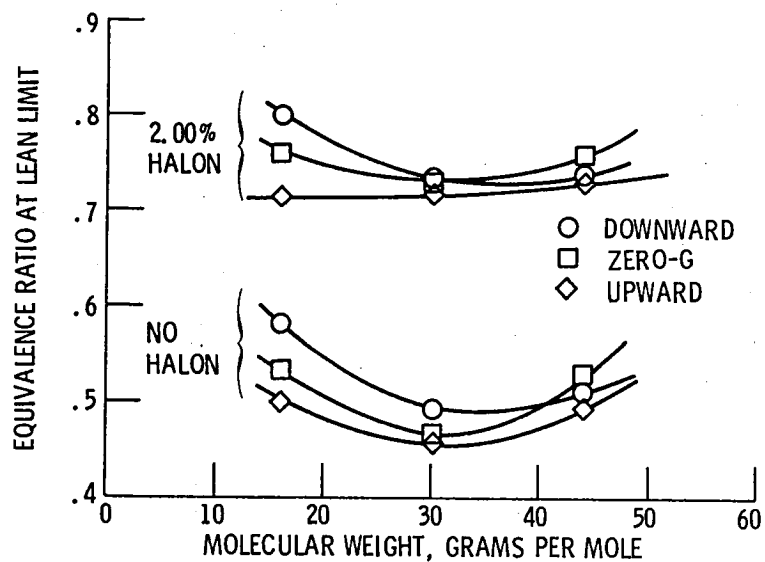


Figure 3. - Effect of fuel molecular weight on flammability limits in uninhibited and inhibited flames at 760 torr initial pressure.

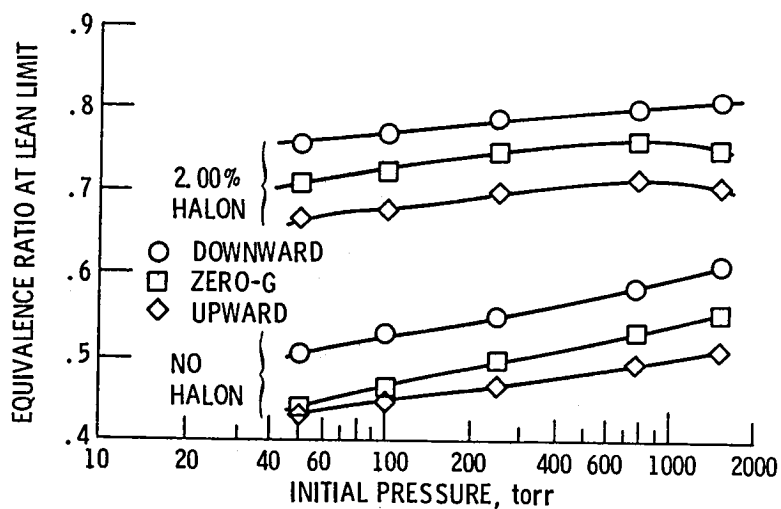


Figure 4. - Effect of initial pressure on methane-air flammability limits in uninhibited and inhibited mixtures.

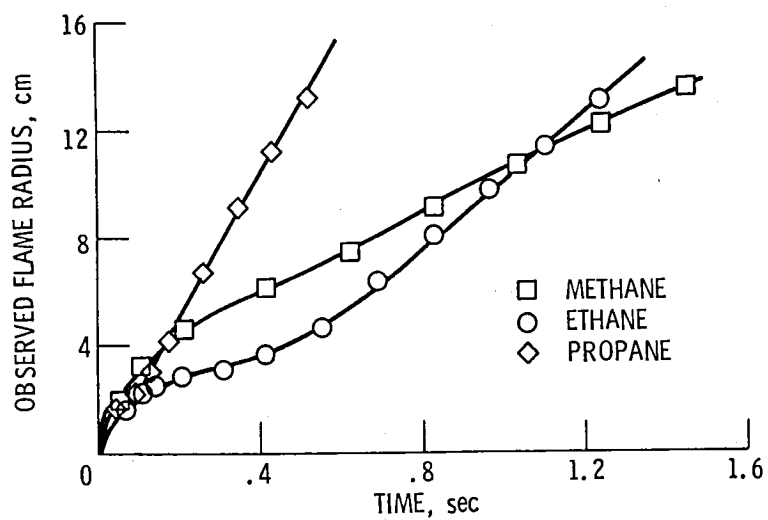


Figure 5. - Effect of fuel type on flame development characteristics in lean limit flames at 760 torr initial pressure.

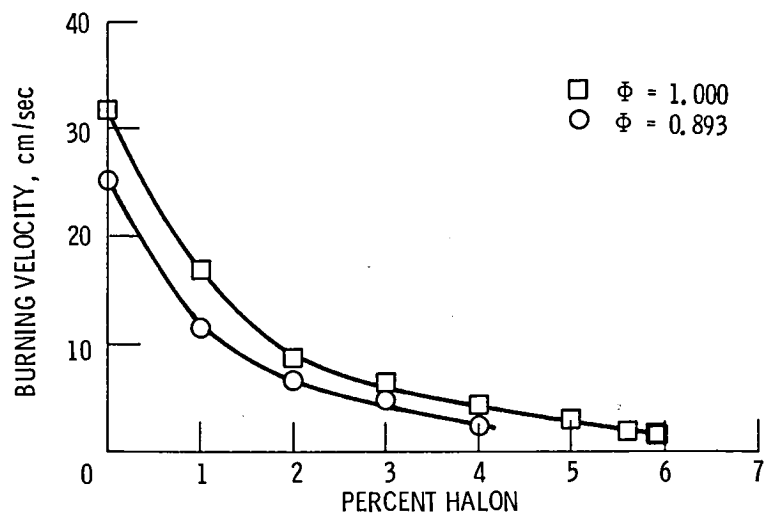


Figure 6. - Effect of Halon concentration on methane-air burning velocities at 760 torr initial pressure.

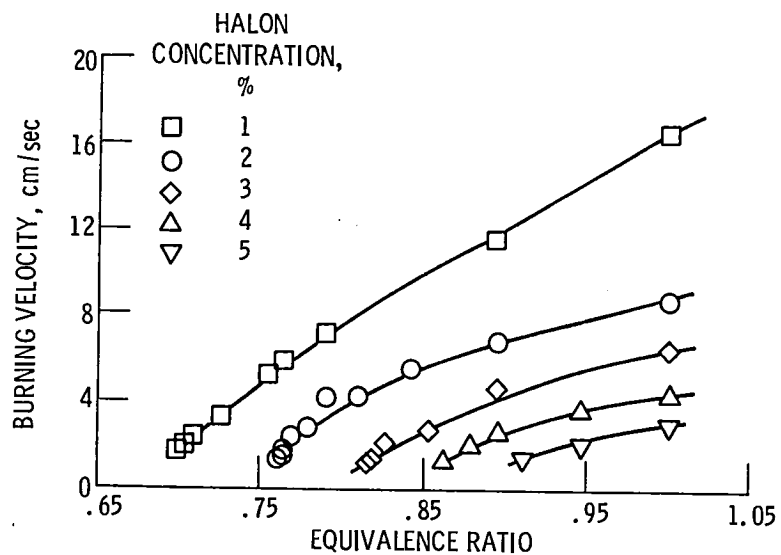


Figure 7. - Effect of equivalence ratio and Halon concentration on methane-air burning velocities at 760 torr initial pressure.

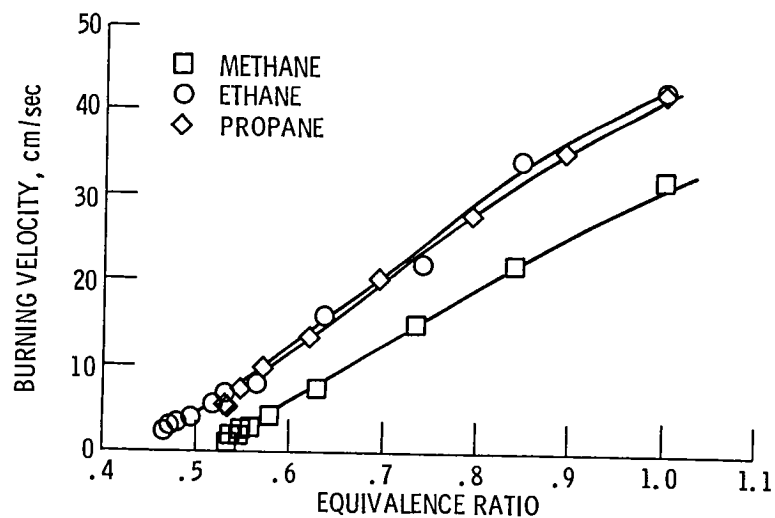


Figure 8. - Effect of equivalence ratio and fuel type on burning velocities in uninhibited flames at 760 torr initial pressure.

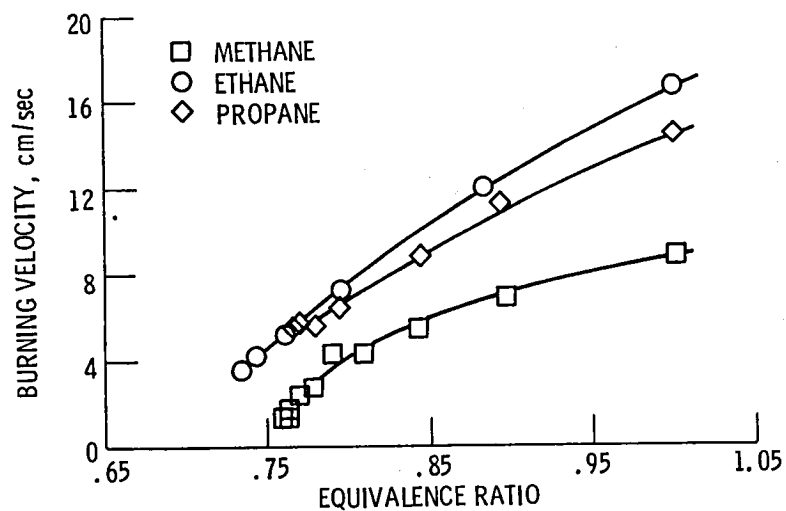


Figure 9. - Effect of equivalence ratio and fuel type on burning velocities in inhibited flames at 760 torr initial pressure.

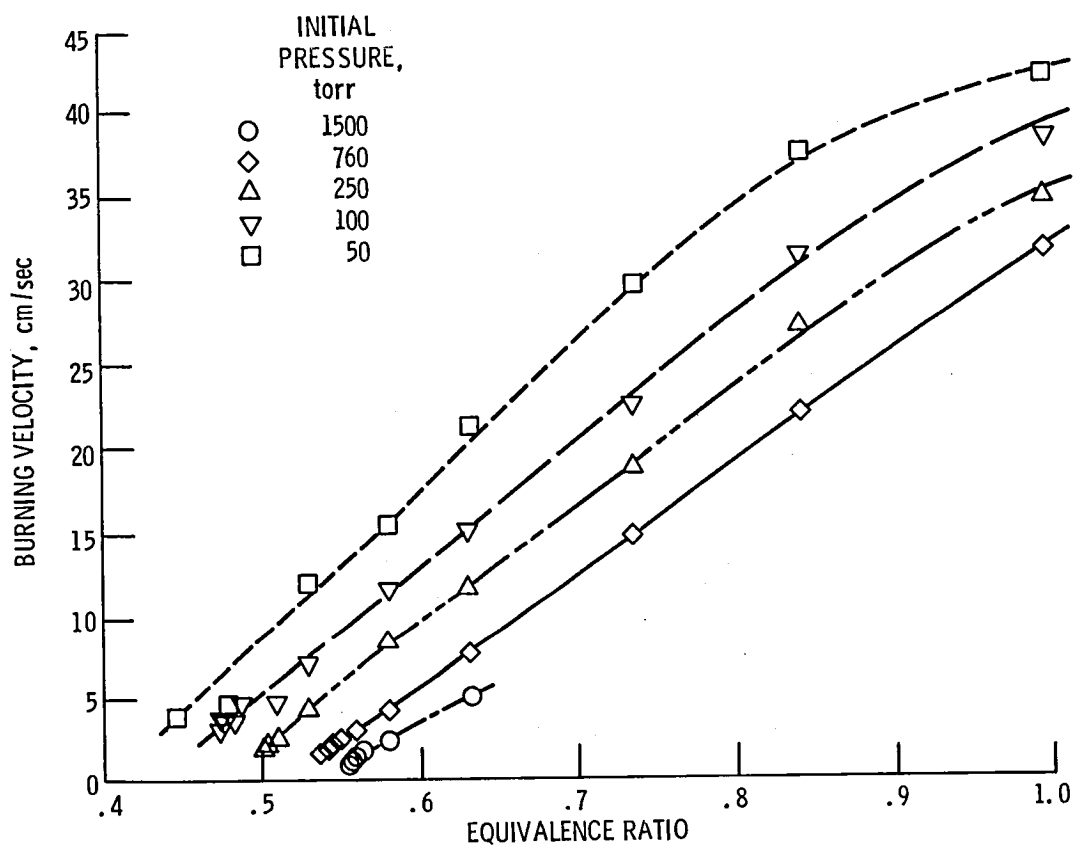


Figure 10. - Effect of equivalence ratio and pressure on methane-air burning velocities in uninhibited mixtures.

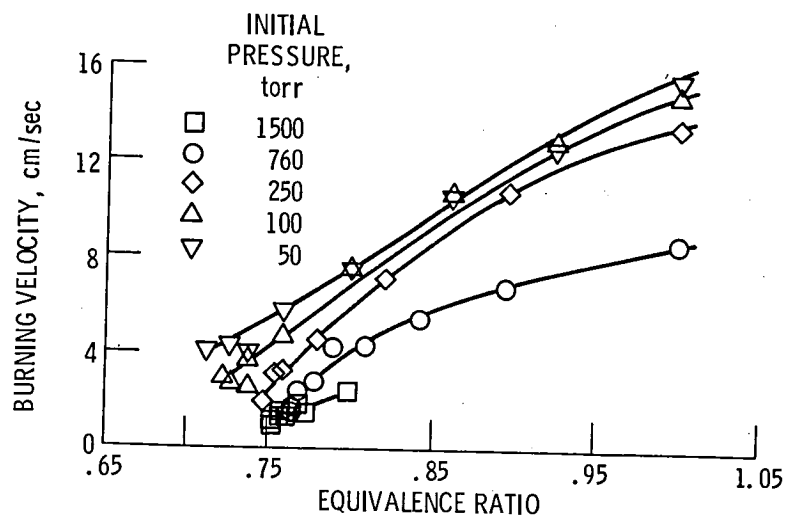


Figure 11. - Effect of equivalence ratio and pressure on methane-air burning velocities in inhibited mixtures.

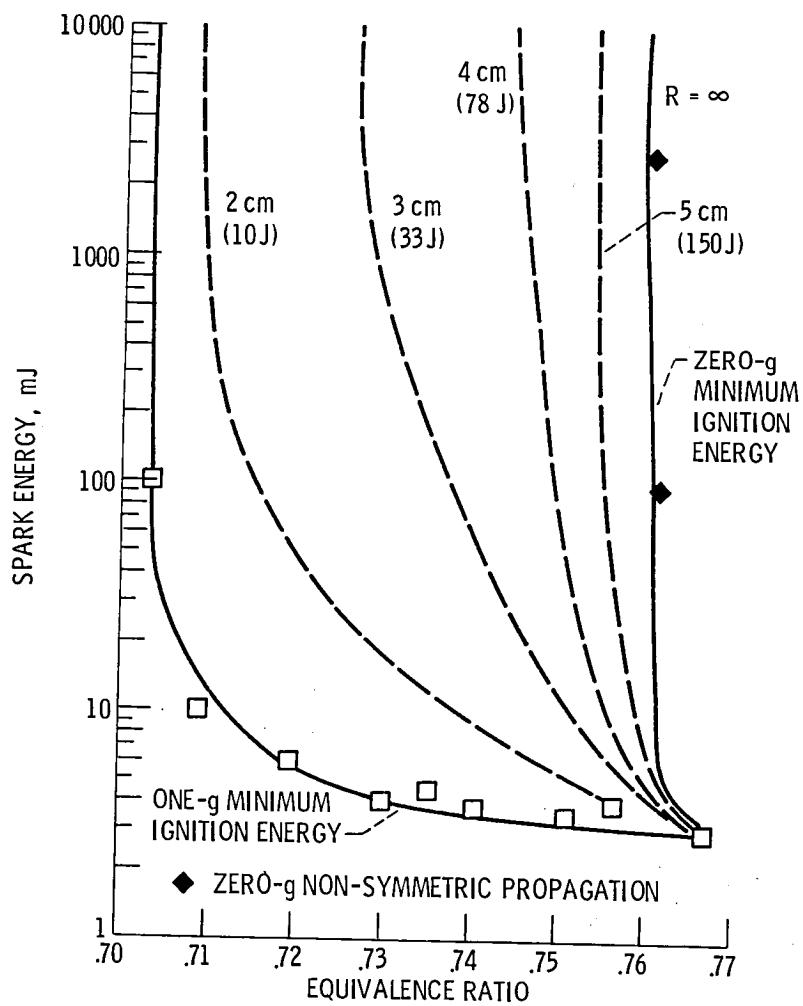
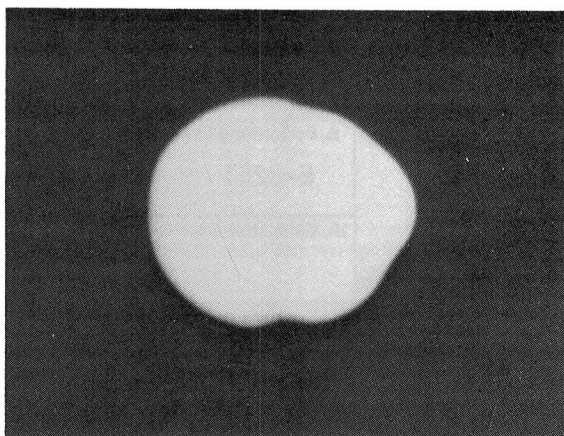
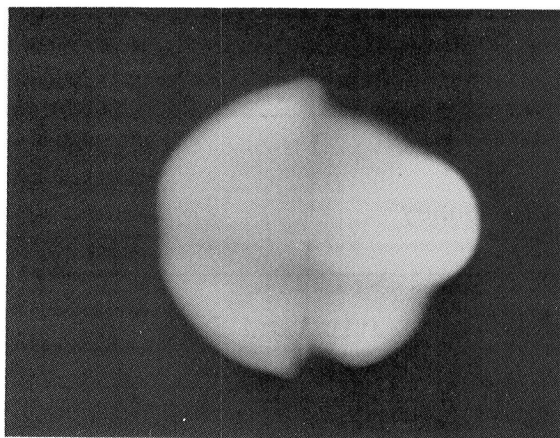


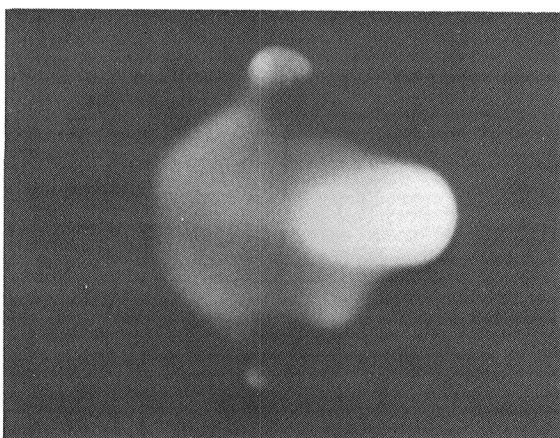
Figure 12. - Effect of equivalence ratio and spark energy on propagation characteristics of inhibited near-limit methane-air flames at 760 torr initial pressure.



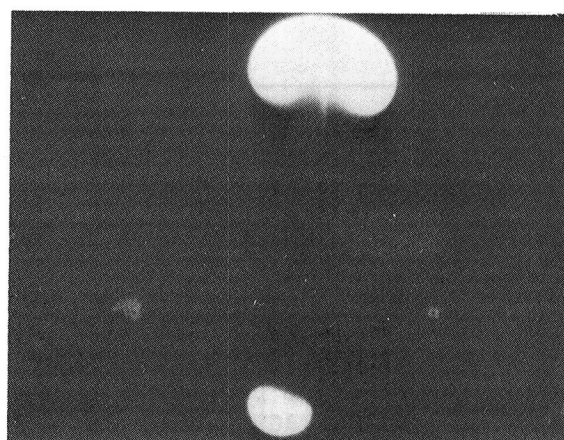
T = .36 sec



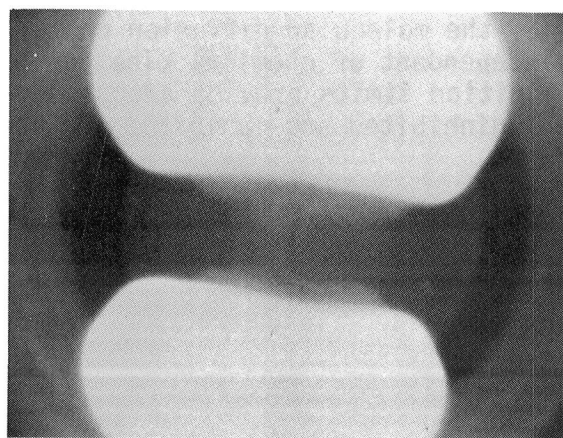
T = .57 sec



T = .72 sec



T = .95 sec



T = 1.48 sec

Figure 13. - Sequential photographs of combustion of mixture of 7.25% methane in air with 2.00% CF_3Br added showing non-symmetric flame propagation at zero-g.

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| 15. Supplementary Notes Paul D. Ronney, NRC-NASA Resident Research Associate. Prepared for the Thirty-fifth Congress of the International Astronautical Federation, Lausanne, Switzerland, October 7-13, 1984. | | | | | |
| 16. Abstract <p>Flammability limits, burning velocities, and minimum ignition energies under initially quiescent conditions were measured for stoichiometric and fuel-lean methane-, ethane-, and propane-air mixtures containing varying concentrations of Halon 1301. The characteristics of near-limit flames were strongly affected by fuel type but not Halon concentration. The conclusions were that the mechanism of the flammability limits was affected by fuel type but not Halon concentration, that the zero-g flammability limit is probably related to a stability criterion which is affected mostly by the molecular diffusion characteristics of the reactant gases and is mostly independent of chemical kinetics, and that the one-g upward flammability and ignition limits provide adequate criteria for safety at one-g and zero-g for both uninhibited and inhibited mixtures.</p> | | | | | |
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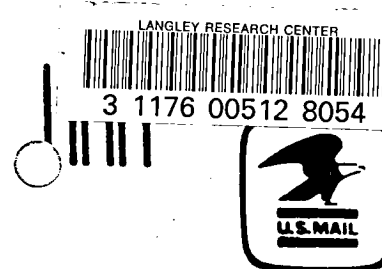
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